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Version of attached file:

Accepted Version

Peer-review status of attached file:

Peer-reviewed

Citation for published item:

Abdelaziz, Moheb and Homaeigohar, Shahin and Hedayati, Mehdi Keshavarz and Assad, Mhd Adel and Elbahri, Mady (2019) 'Solar aluminum kitchen foils with omnidirectional vivid polarizonic colors.', *Advanced optical materials.*, 7 (15). p. 1900737.

Further information on publisher's website:

<https://doi.org/10.1002/adom.201900737>

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Solar Aluminium Kitchen Foils with Omnidirectional Vivid Polarizonic Colors

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Keywords: ((solar absorber, plasmonic dipoles, structural coloration, polarizonic effect)).

Abstract: To meet the need to the “customized eco-friendly” design of flexible solar materials in vivid colors, here, the solar kitchen foils are devised which are selective, omnidirectional and colored. The experimental and simulation results enable drawing the roadmap of the fabrication of glowing, colored flat foils for diverse energy, packaging and decoration purposes. A new gold free golden foil, i.e. mimicking gold optically and visually, is designed as a prototype for the sustainable fabrication of advanced colored foils with glowing colors e.g. golden with no need to neither hazardous anodization nor dying processes. The solar foil

performs based on the newly developed concept of the polarizonic interference allowing production of omnidirectional structural colors by a disordered plasmonic nanocomposite. As the specific highlight, selective reflective coloration by plasmonic dipoles in a hybrid dielectric host, i.e. the building block of ultrathin solar absorbers with tailored, vivid colors, on an aluminium foil is demonstrated. In terms of the production technique, the applied sputtering technique is simple, versatile, cost-effective, and compatible with the industrial packaging, decoration and solar absorber manufacturing processes. Thus, it holds great promise for creation of advanced, flexible, colored solar absorbers in a simple, scalable, and sustainable fashion.

1. Introduction

Solar materials are the materials able to selectively absorb, reflect, and transmit the inexhaustible solar radiation at the optical frequencies and to be integrated in the green energy saving applications. In the last decade, a large amount of investment and attention has been paid to produce cheaper solar based devices. However, there are still challenges such as the sustainable manufacturing techniques, materials cost, and flexibility of solar materials among others ^[1].

Aluminium foil especially in metallic vivid colours has been used for art, decoration and packaging. With respect to energy applications, it is indeed an interesting industrial solar material thanks to its lightweight, high strength, optimum surface reflectivity, and excellent electrical and thermal conductivities. Aluminium is being used as back reflector and contacts in solar cells because of its abundance, low cost and compatibility with silicon technology and even more as building block of the solar absorber over the past decades ^[2, 3]. The coloured aluminium is made through an anodizing process with or without dyeing i.e. inclusion of artificial nanoparticle pigments. By the anodizing process, a porous aluminium/ aluminium

oxide layer with various pore sizes hence distinctive colors is realized ^[4, 5]. During the process, the surface can be transformed from a reflector to an absorber by controlling the exposure time to electrolyte. Also, by electrodeposition of metal nanoparticles made of iron, nickel, and/or of pigments, a solar absorber is realized. Despite the simplicity of the anodization process, its implementation for construction of the solar aluminium foil is difficult, as the foil is fragile and mechanically weak. Most importantly, the process is problematic in terms of pollution and involvement of hazardous chemicals. As an extra burden to environment, the fossil fuel related pollution and chemical wastes must be addressed. Particularly, the hazardous byproducts resulted from the dissociation of diverse chemical compounds are challenging. The flat coating process enabling production of shiny and matt coloured materials based on metallic foils can be a great alternative method that minimizes the environmental concerns. However, the design of a selective solar aluminium foil with controlled reflectivity and absorptivity is tricky and achievable generally by interference-based multilayer thick coatings in compliance with the quarter-Lambda rule. In such a structure, optical properties vary at different incidence angles (e.g. during the day) because the light transmission through the stacks is angular and/or polarization sensitive^[6]. To address this shortcoming and to endow the solar foil materials with angle and polarization independency, there is a need to innovation in the design of such solar energy materials.

Here, we take the first step to overcome the aforementioned problem and to initialize production of a new generation of efficient and cheap energy kitchen foils. This ambition can be realized by advancement of nano-optics through engineering of color designs. Here, we devise a new class of the polarizonic optical systems with unprecedented control of light and color. We introduce the polarizonic interference phenomenon as a new design tool for artificial

structural colours of disordered dipoles with unique metallic, matt and shiny appearance that are omnidirectional and irrespective to the polarization/angle of the incident light.

As a proof of concept, first a gold free golden foil is constructed that can exactly mimic the optical response of gold and hence appear as golden. This golden colored Al foil could be suggested for a diverse range of decorative applications that need a flexible, robust and eco-friendly coating. As the second, more sophisticated utility, the first solar foils with matt to shiny color gradients are introduced by controlling the type of the nanoparticulate inclusions and refractive index of the hybrid host. In such solar foils the absorption is preserved above the agreed term of perfect absorption i.e. 90% while vivid colors also emerge.

The Polarizonic Interference Effect, towards a Gold Free Golden Foil: It has been thought that the shiny and vivid colors are solely achievable by the optically symmetric and periodically structured materials ^[7-10], which is "*not quite right*", based on our recent progresses in the field. Several disordered structures consisting of nanoparticles and molecules that are able to show coherent and cooperative specular reflection "polarizonic" responses have been recently introduced by us^[11-15] and implemented for creation of vivid colors. Despite various merits with respect to structural polarizonic coloration, this mode of specular reflected colour is angular and polarization dependent thus inefficient for different applications such as optical measurement systems, colored display devices, and solar absorbers.

Here we propose and validate a concept whereby the polarizonic based coloration can be arisen in an omnidirectional mode. To commence, we demonstrate that generation of vivid, shiny colors is not pre-conditioned by absolute blockage of the reflection and a non-distinct or "rest" reflection spectrum should arise. The color of gold is a unique instance to base the concept on and understanding the gold-light interaction at the atomic scale can enable us to

realize a similar optical response by the gold free components even with an omnidirectional response.

The classical free electron theory describes the reflectivity of metals as a continuous field whose intensity depends only on the number of free electrons per unit volume and hence conductivity. Therefore, different metals offer a broad band reflection in the IR and Visible frequencies. While this view describes the grey appearance of some metals and even semi-metals (Al, Ag, and Si) well, it fails to describe the golden color of gold, for instance. The classical Drude-Lorentz theory identifies a discontinuity at the surface, where the component of electric intensity perpendicular to the surface fluctuates extensively in a transition layer (i.e. light penetrates into a metal only at a short distance). Accordingly, as described by the Lorentz classical harmonic oscillators, a localized state is assigned at the longitudinal plasma frequency. In the simplest localized plasmonic case, the alternating surface charges form an oscillating dipole that radiates electromagnetic waves to the far field^[12]. This simple model for the dipole is evocative of an optical antenna and well known as the electronic polarization occurring at the optical frequencies that causes a local reflection what we call “polarizonic”. Accordingly, the metal behaves as a series of classical free electrons and classical harmonic oscillators with a characteristic broad reflection of the free electron “continuous” and a localized reflection “discontinuous” at the plasma frequency^[16]. The existence of both the states in the metal is the pre-condition of formation of a quantum interference that governs the drop of reflection at the plasma frequency, **Figure 1a**. In the case of gold, it occurs at the blue range of visible frequencies (the wavelengths of 440-490 nm), hence giving rise to the golden color. However, as the reflection does not drop to zero, the metal appears shiny. It is essential to note that, absence of the localized reflection of plasma oscillation at the visible frequency results in the grey color of metals like aluminium and silver. Based on these facts, it can be deduced that *whenever a localized state of “discontinuous” polarizonic reflection of plasmonic*

nanoparticles meets a “continuous” broadband reflection of a grey metal or a semi-metal, a polarizonic interference emerges, Figure 1a. To validate this postulate, an aluminum foil and some other grey appearing substrates (stainless steel and silicon) are coated with a silver nanocomposite layer (as thin as 20 nm, and with 33% Ag). This combination generates a polarizonic interference at the blue range of visible frequencies (where the nanocomposite reflects light), Figure 1b, thus induces an optical response equal to gold's and a similar golden appearance to the foil and stainless steel (Figure 1c) even with no presence of gold as a constituent material.

Interestingly, the golden color of the foil can be controlled by the filling factor of the nanocomposite. For instance, employing a nanocomposite overlay containing 20% Ag nanoparticles endows the foil with a titan silver color, Figure 2a. The titan color arose by the reflectivity drop at 400 nm, Figure 2b. The simulation match reasonably with measurements as shown in Figure 2b.

The shiny foil is omnidirectional in fact. Figure 2c–e shows the total and polarized polarizonic reflection properties of a 20 nm Ag nanocomposite film/Al foil under oblique incidence. Apparently the minima at 400 nm is almost stable when the incidence angle rises from 45° to 75°. To illustrate the phenomenon, the polarized reflection (under s- and p-polarized light illumination) is considered. The results stress on the polarization/angle independency of the polarizonic interference effect. It is worthy to note that the intensity difference between the simulation and experimental data arose from the fact that the simulation is based on pure Al as the backplane while the experiments are based on the kitchen aluminum foil (which is not as pure as that assumed in the simulation).

We believe that the dipolar destructive interference plays a key role in the polarizonic interaction, as clearly shown in Figure 1b. The finite difference time domain (FDTD) simulations witnesses this effect as well. The FDTD simulation of the 20 nm Ag

nanocomposite, whose details have been described elsewhere^[17], can verify the striking properties of the cooperatively interacting ultrafine particles and their interaction with a conductor. As seen in Figure 2f, an ensemble of ultrafine (5 nm) Ag nanoparticles in a thin dielectric matrix brings about electromagnetic confinement and guiding as well as an unexpected spatial radiated field. This radiation in the Z direction of ultrafine particles differs strongly from the modes of dipolar coupling in the X direction (hot spots). The electromagnetic fields are weakened if the nanocomposite overlays a semi conducting surface of e.g. silicon, Figure 2g. The dipolar interference of the plasmonically excited electric dipole and its image i.e. the induced opposite dipoles on the conductor forms a non-radiative quadrupole mode giving rise to an anti-reflection event and governs the design of omnidirectional polarizonic interference.

Plasmonic Polarizonic Reflection in a Hybrid Dielectric: to tailor the range of the operation frequency wherein the polarizonic interference emerges, the polarizonic reflection resonance must be adjustable. The plasmonic nature of majority of metamaterials allows to adjust their resonance peak by changing the size, shape, type and the surrounding environment of their metallic constituent (i.e. via dielectric changes). The limited number of dielectrics challenges the convenient design of the resonance band in a wide range of frequencies. Here, we tune the resonance by developing a hybrid dielectric whose refractive index can be adjusted by variation of the ratio of the components instead of changing the type of the surrounding. Accordingly, a strong tunability of the reflection band is obtained in an ultra-thin nanocomposite reflector composed of disordered gold-silica (SiO₂) -titania (TiO₂) nanocomposite, only by slight changes in the composition of the hybrid dielectric. The TEM micrograph of the SiO₂-TiO₂ hybrid film is shown in Figure 3a. The hybrid film is amorphous due to immiscibility of SiO₂ and TiO₂^[18-21]. The advantage of such an amorphous structure is its isotropic optical

performance that limits the whole structure's optical behaviour to merely the nanoparticles'. The ellipsometry analysis followed by modelling allows us to determine the refractive indices (RIs) of the hybrid film, Figure 3b. It turns out that, there is an almost linear relationship between the ratio of the oxides and the RI of the hybrid film. The RI of the film can rise from 1.65 up to 2.2 by increase of the ratio of $\text{TiO}_2/\text{SiO}_2$ from 30 to 50%. It is well-known that the plasmon resonance of particles strongly depends on the type of their surrounding environment. To assess the extent of the resonance shift by environmental changes, we studied the polarizonic reflection spectra of the Au-SiO₂-TiO₂ composite deposited on a glass substrate. The Au volume fraction was 40% while the ratio of $\text{TiO}_2/\text{SiO}_2$ was changed. A TEM image of the nanocomposite, Figure 3c, clearly shows the random distribution of the closely spaced particles in the matrix. Figures 3d-i imply the p- and s-polarized reflection of the Au nanocomposite (40% Au NP) with a dielectric and a hybrid dielectric matrix of SiO₂ and TiO₂. According to the presented graphs, the RI increases when silica is partly or wholly replaced by titania and hence the polarizonic peak is red shifted. Additionally, the light reflection is intensified. Most interestingly, the Brewster wavelength is red shifted relative to the one appearing at the Brewster angle under the p-polarized light. The Brewster wavelength effect, we introduced recently ^[11, 12] as a means for sensing the net polarity of a plasmonic composite, acts through the dipolar interaction between the excited dipoles and the host matrix. Such an interaction governs a net dipole moment at the interface based on the interfacial polarizability effect^[22].

A vivid colored solar absorber: The polarizonic interference effect introduced here can be regarded as the basis of a new generation of bilayer solar perfect absorbers. It is worthy to note that, the traditional perfect absorbers typically comprise three layers of reflector, dielectric spacer, and nanocomposite^[23-25], and because of such a structure look totally black. In contrast, here a bilayer perfect absorber in diverse colors is generated by the polarizonic interference

that can be tuned by the type and the filling factor of the plasmonic nanomirrors and dielectric hosts. As a prototype for this concept, here, we design an inexpensive, color tailored solar foil via coating of a kitchen Al foil with an ultrathin layer of Au nanocomposites containing different Au nanoparticle filling factors and various dielectric hosts. Au was implemented for the design of such a perfect-colored absorber based on our previous studies on both well-known absorbing metals of Au and Ag.^[12] Given the optical constants (including RI (n) and extinction coefficient (k)) of both the metals in the visible range, the intrinsic absorption of gold is much higher than that of silver, and for this reason it was selected for the construction of the solar absorber. Deposition of the Au/SiO₂ nanocomposites on an optically thick Al foil (≈ 0.01 mm) generates perfect absorbers with various colors (Figure 4a). Depending on the Au filling factor, the emerged color varies from rose red (20% Au), wine red (25% Au), purple (32% Au) to green (40% Au). In case the Au filling factor is kept at 40% and the host matrix changes to TiO₂ or SiO₂/TiO₂ (50/50), the color alters to golden and brown, respectively, as shown in Figure 4a. Figure 4b shows the absorption/reflection spectra of this coating when the Au filling factor and the dielectric host changes. An obvious broadening and shifting of the spectra to longer wavelengths occur by insignificant changes of the Au filling factor and the dielectric hosts' type. The emerged vivid color is determined by the reflection and interference responses, as illustrated in the inset of Figure 4b. Such major changes in the spectra (in terms of bandwidth and maximum intensity) witness the notable tunability of the coating for the design of perfect colored absorbers among other optical applications. Thus, a coating material with strong interference at the visible wavelengths is formed that acts independent to the reflectivity of the underlying foil. In fact, the performance of the coating in terms of color generation is totally dependent on the nanocomposite overlay. For instance, coating of the foil with the plasmonic nanocomposites containing 40% Au with different dielectric hosts of TiO₂ and SiO₂ leads to emergence of shiny and matt colored perfect absorbers, respectively. The RI contrast between

the host material i.e. TiO_2 and SiO_2 is responsible for generation of the mentioned shiny color. In fact, the reflective, shiny appearance of the composite is caused by the higher RI of TiO_2 which is not the case for the composite with the SiO_2 host, appearing matt.

2. Conclusion

Taking together, the flexible aluminium foils in golden and vivid colors were designed and demonstrated based on the polarizonic interference effect. The presented concept could enhance our understanding of the dipolar reflection and the interference mode and potentially will play a pivotal role in the development of a new generation of colored solar perfect absorbers. Such solar absorbers can be based on inexpensive, flexible thus easily shapeable Al kitchen foils thus hold a great promise for scalable, economical production of this sort of solar devices. As the fabrication technology i.e. sputtering is an industrially established method, the proposed method offers interesting potentials for designing perfect-colored absorbers.

3. Experimental Section

Fabrication of the solar kitchen foils: All the films comprising the nanocomposite overlaid Al foils were synthesized by a magnetron sputtering machine designed and assembled in house. In this machine, the homemade metal vacuum chamber was first evacuated to below 10^{-6} mbar. In this magnetron sputtering system, two magnetrons can be run independently and a sample holder is attached to a rotatable motor to provide a uniform film during deposition and to avoid a gradient in the composition and thickness. Such a set-up enables synthesis of nanocomposite films with various metal filling factors so that via a RF magnetron source, the dielectric is sputtered and via a DC magnetron source, the metal nanofillers. These magnetrons

are placed in opposite directions relative to the sample holder and at a 50° angle relative to the substrate.

Characterizations: The filling factor of the nanocomposite was estimated based on the rate of deposition. The UV/vis/NIR spectrometry (LAMBDA900-PerkinElmer) was used to measure the reflectivity and transmission of the samples. The film thickness measurement and ellipsometry analysis of the dielectric films (hybrid samples) were carried out by an automated angle M-2000® Ellipsometer (J. A. Woollam Co.). For modelling of the optical properties, the software provided by the company (CompleteEase) was used. In order to model the refractive index of the hybrid films, the Maxwell-Garnet theory was employed. A profilometer (Dektak XT-E-BRUKER) was also applied for mechanical thickness measurements. TEM analysis of the samples was done with Tecnai F30 G² - ST. For this sake, the films were deposited on “carbon coated copper grids”.

COMSOL Multiphysics was used for simulation of the reflection of the nanocomposites. The silver refractive index (imaginary part) was multiplied by 1.5 in order to include the effect of larger extinction of small nanoparticles. The unit cell of the simulation was set to be 30x30 nm and the incident light was linearly polarized in X direction. The nanocomposite was set to be made of 4 layers and the nanoparticles were assumed to be 4.4 nm in diameter. The type of the periodicity was set to be continuity. A perfectly matched layer (PML) layer of 100 nm thickness was applied below the metal layer.

Acknowledgement

M.E. thanks the School of Chemical Engineering at the Aalto University, Finland, the Helmholtz Association, Germany, for providing financial support to his research group. The authors gratefully acknowledge partial financial supports from the Academy of Finland and the German Research Foundation (DFG) through SFB677 (C01).

Conflict of Interest

The authors declare no conflict of interest.

Received: ((will be filled in by the editorial staff))

Revised: ((will be filled in by the editorial staff))

Published online: ((will be filled in by the editorial staff))

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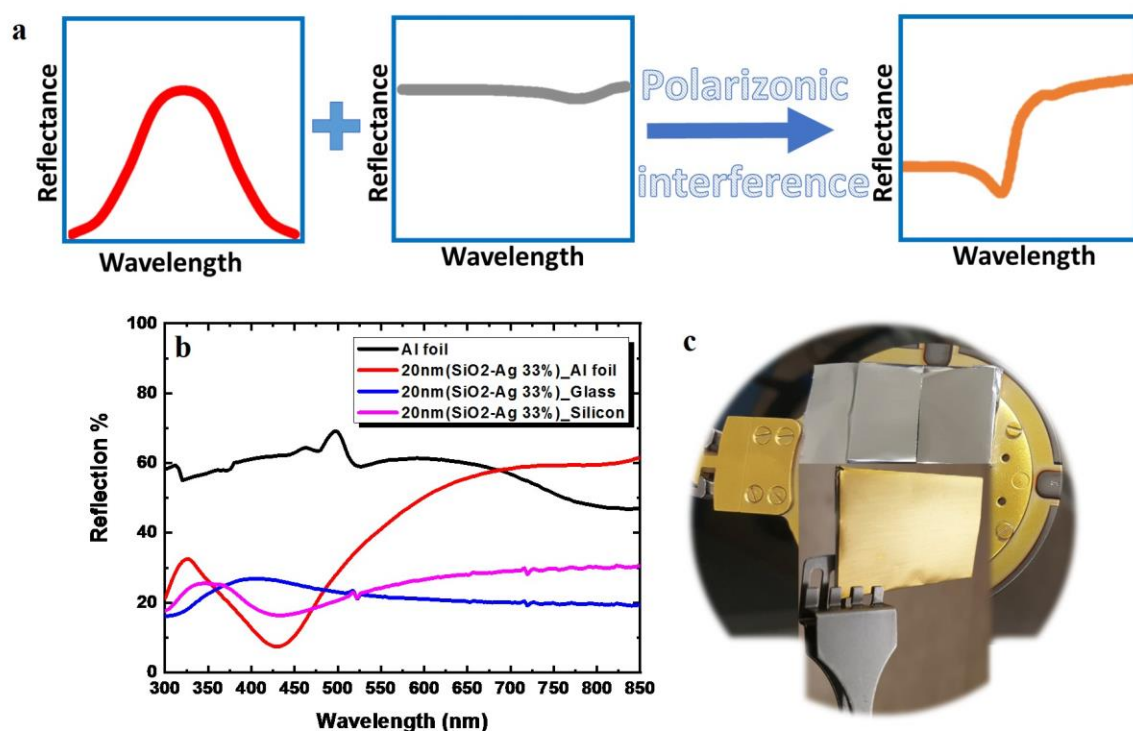


Figure 1. The polarizonic interference concept. a) Schematic illustration of the destructive interference of the specular reflection with the broad reflection of a conductor. This destructive interference leads to a reflectivity drop at the resonance frequencies. b) The polarizonic reflection of a Ag nanocomposite (33% Ag) on glass appears at the blue range. To realize the polarizonic interference and thereby a reflectivity drop, an Al foil and/or Si layer is overlaid by the nanocomposite. c) Overlaying the nanocomposite on grey substrates (Al foils, and stainless steel.) brings about a golden color through the polarizonic interference effect.

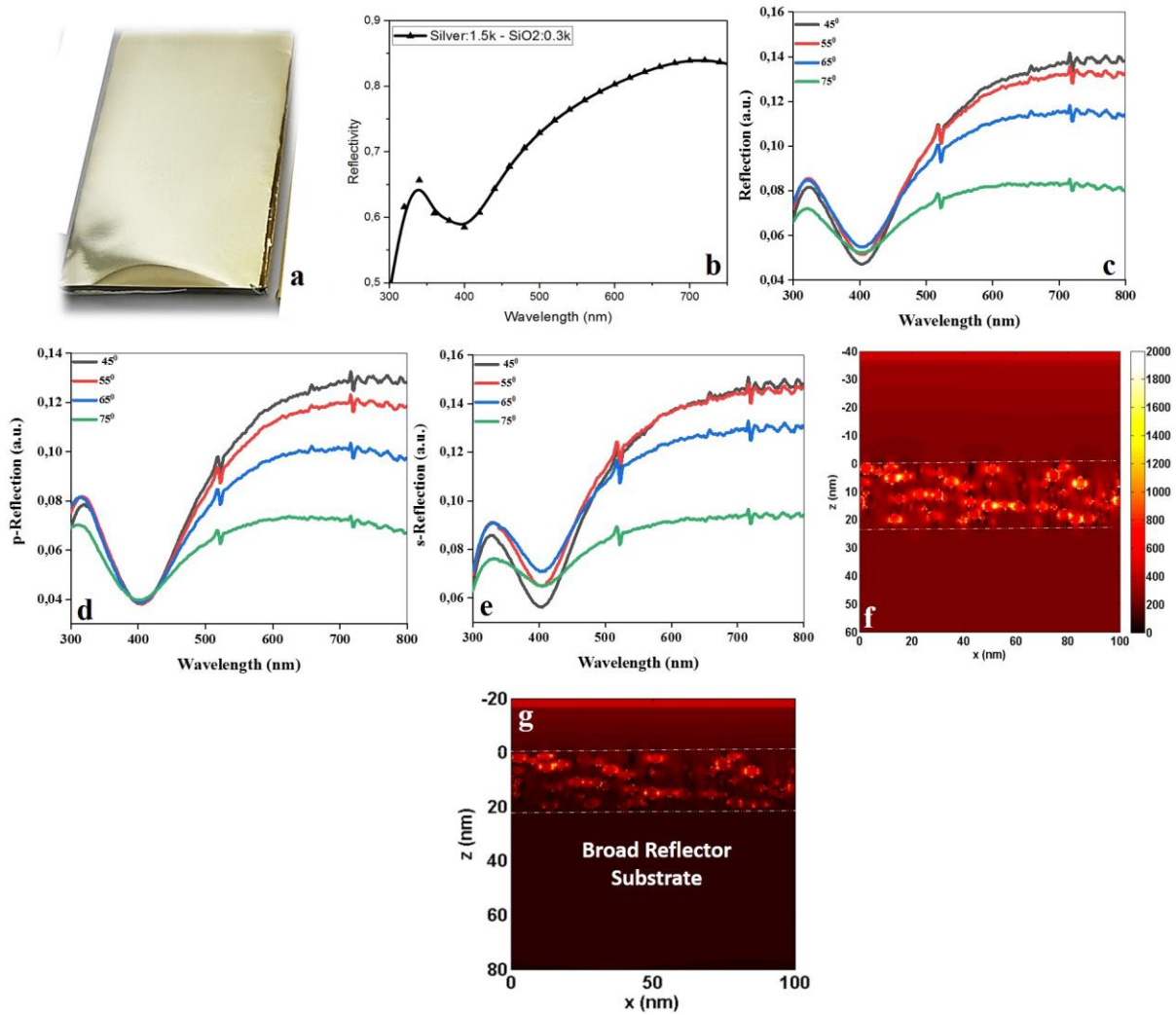


Figure 2. The angle independency of the polarizonic interference based color. a) Coating an Al foil with a Ag nanocomposite (20% Ag) overlay brings about a titan color for the foil through the polarizonic interference effect. b) Reflectivity spectrum for the 20 nm Ag/SiO₂ nanocomposite. c-e) The total, p-polarized, and s-polarized polarizonic reflection spectra of a 20 nm Ag/SiO₂ nanocomposite/Al foil hybrid structure at various oblique incidence angles. The FDTD simulation uncovers the electrical field amplitude distribution across the cross section of a Ag/SiO₂ nanocomposite (containing 20% of arbitrarily arranged separate silver nanoparticles as small as 5 nm in a generic monodisperse dielectric matrix ($\epsilon = 2.25$)) as alone (f) and as deposited onto a grey substrates, here silicon substrate (g). Note that an incident light with the wavelength of 430 nm has been regarded in this study.

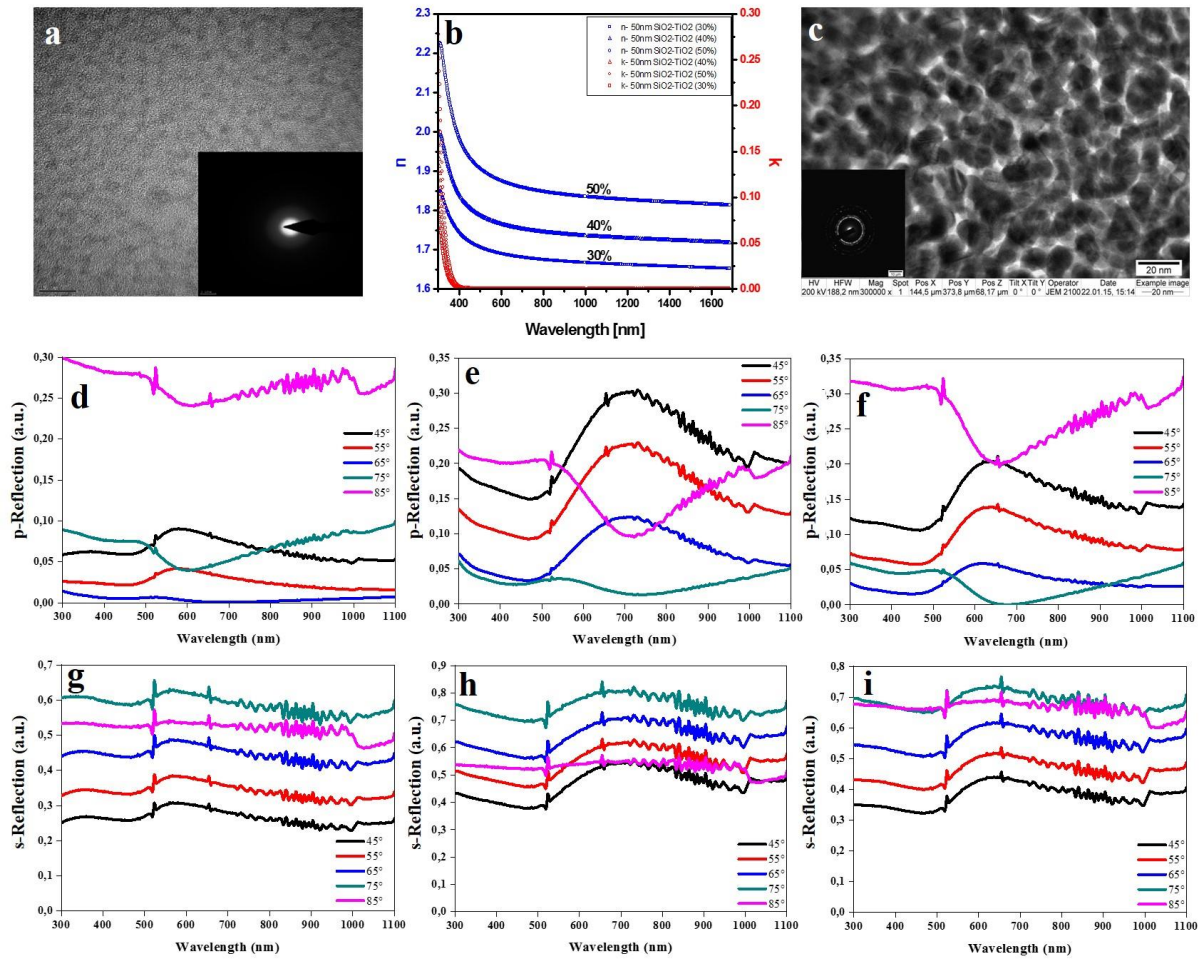


Figure 3. Tuning the polarizonic reflection via a hybrid dielectric. a) TEM image shows the hybrid film composed of TiO_2 - SiO_2 . The inset image represents the corresponding diffraction pattern of the hybrid dielectric. b) RI dispersion of the SiO_2 - TiO_2 hybrid film with different ratios of the consisting oxides, calculated by the Maxwell-Garnet modelling of the ellipsometry data. The blue and red curves represent the real and imaginary part of the RI, respectively. c) A typical top-view TEM image of the Au nanocomposite (40% Au)-hybrid dielectric (50/50). The inset shows the diffraction pattern of the structure. d&g) p- and s-reflectivity of 40% Au NP/ SiO_2 , respectively. e&h) p- and s-reflectivity of 40% Au NP/ TiO_2 respectively. f&i) p- and s-reflectivity of 40% Au NP/ (50/50) SiO_2 - TiO_2 , respectively.

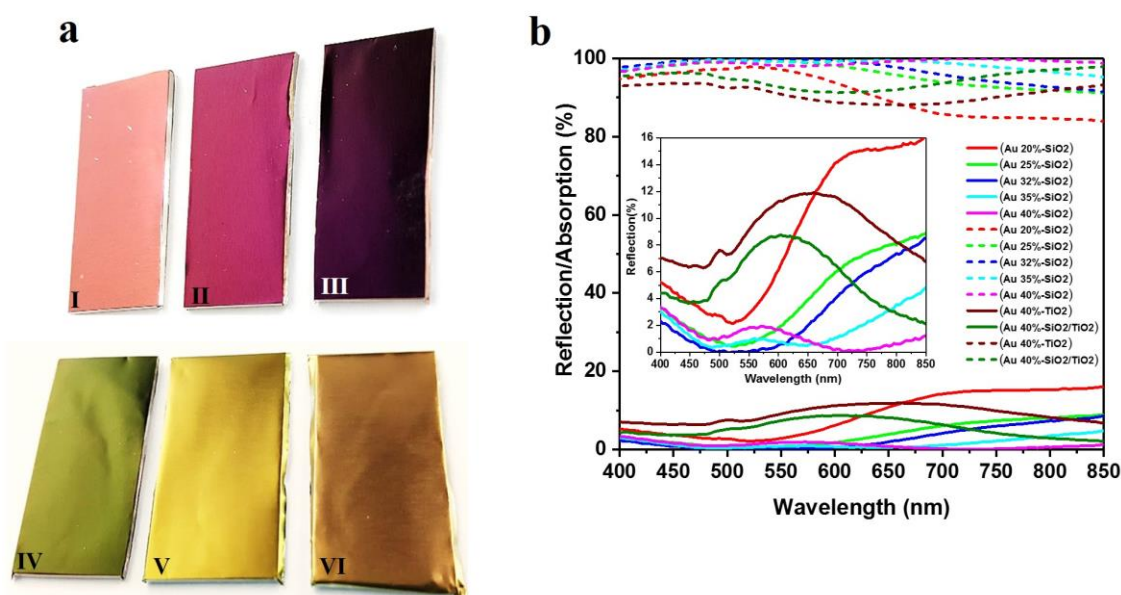


Figure 4. Perfect colored absorption by the polarizonic gold nanocomposites on an aluminum foil. a) The perfect absorbers based on the (Au/SiO₂)/Al layered structure with various colors ranging from rose red (I; 20% Au/SiO₂), wine red (II; 25% Au/SiO₂), purple (III; 32% Au/SiO₂) to green (IV; 40% Au/SiO₂), respectively. By replacing the SiO₂ host with TiO₂ and a hybrid thereof, the colors change to golden (V; 40% Au/TiO₂) and brown (VI; 40% Au/(TiO₂/SiO₂), respectively. In the latter composition, the amount ratio of SiO₂/TiO₂ is 50/50. b) The absorption/reflection spectra of the aforementioned absorber when the Au filling factor and the dielectric host change. The inset Figure shows that the emerged vivid colors are determined by the reflection and interference response of the absorber with respect to its varying composition.

The aluminum kitchen foil based flexible solar materials are produced in vivid colors in a “customized eco-friendly” manner. These colored solar foils are selective and angle independent. The solar foil’s performance mechanism is based on the new concept of the polarizonic interference that enables production of omnidirectional structural colors by a disordered plasmonic nanocomposite overlaid an aluminium foil.

Keyword ((Solar colored kitchen foil))

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ToC figure

